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Contents

	Page
Thermodynamic Properties of Argon in the Temperature Range -100 to +600° C. and Pressure Range 0 to 80 Atmospheres— <i>E. Whalley</i> - - - - -	111
A High-temperature X-ray Goniometer— <i>R. G. Butters and J. Gordon Parr</i> - - - - -	117
The Penetration of Concrete by Radionuclides in Solution— <i>L. H. Gervantman and D. Sam</i> - - - - -	122

CANADIAN JOURNAL OF TECHNOLOGY

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THERMODYNAMIC PROPERTIES OF ARGON IN THE TEMPERATURE RANGE -100 TO $+600^{\circ}\text{C}$. AND PRESSURE RANGE 0 TO 80 ATMOSPHERES¹

By E. WHALLEY

ABSTRACT

The values of PV in the temperature range -100 to $+600^{\circ}\text{C}$. and specific heats, enthalpy, entropy, and Joule-Thomson coefficient, in the temperature range 0 to 600°C ., have been calculated for argon in the pressure range 0 to 80 atm. using the compressibility data presented in a previous paper.

In a previous paper (4) the measurement of the compressibility of argon in the temperature range 0 to 600°C . and pressure range 10 to 80 atm. was reported. The values obtained were compared with values reported by previous workers and weighted means of the virial coefficients for the equation

$$PV = A + BP + CP^2 + DP^3 + \dots$$

were taken to obtain the "best" values. The values of B over the temperature range -100 to $+600^{\circ}\text{C}$. and of C over the temperature range 0 to 300°C . were fitted to series equations.

PV for argon in the temperature range -100 to $+600^{\circ}\text{C}$. and a number of thermodynamic properties in the temperature range 0 to 600°C . and pressure range 0 to 80 atm. have been calculated from the data of Ref. (4). The zero pressure properties were obtained from the NBS-NACA Tables of the Thermal Properties of Gases (2) or by simple calculation therefrom. The temperature derivatives of B and C were calculated from the series equations presented in Ref. (4). Below 0°C . there are only two values of the third and fourth virial coefficients so it is impossible to obtain accurate values of their first temperature derivatives, and impossible to obtain any values of the second derivative. Thermodynamic properties which depend on the values of these derivatives cannot be calculated at all accurately and no values are given below 0°C .

The various quantities were calculated as described in a previous paper (1). The Joule-Thomson coefficient μ was calculated from the relation

$$\begin{aligned}\mu &= -(\partial H/\partial P)_T/C_p, \\ &= -[V - T(\partial V/\partial P)_T]/C_p, \\ &= -[(B - TdB/dT) + P(C - TdC/dT) + P^2(D - TdD/dT)]/C_p.\end{aligned}$$

¹Manuscript received November 18, 1954.

Contribution from the Division of Applied Chemistry, National Research Council of Canada. Issued as N.R.C. No. 3484.

TABLE III
CONSTANT VOLUME SPECIFIC HEAT C_v/R FOR ARGON

$T, ^\circ\text{C.}$	0	25	50	75	100	125	150	174	200	300	400	500	600
P atm.													
0	1.5000	1.5000	1.5000	1.5000	1.5000	1.5000	1.5000	1.5000	1.5000	1.5000	1.5000	1.5000	1.5000
1	1.5008	1.5005	1.5004	1.5004	1.5004	1.5003	1.5003	1.5003	1.5002	1.5002	1.5002	1.5002	1.5002
2	1.5015	1.5012	1.5010	1.5009	1.5007	1.5006	1.5006	1.5005	1.5005	1.5004	1.5004	1.5003	1.5003
5	1.5044	1.5033	1.5029	1.5027	1.5022	1.5021	1.5021	1.5021	1.5021	1.5021	1.5021	1.5021	1.5021
10	1.5077	1.5065	1.5061	1.5059	1.5056	1.5053	1.5053	1.5053	1.5052	1.5052	1.5052	1.5052	1.5052
20	1.5111	1.5099	1.5094	1.5091	1.5088	1.5086	1.5086	1.5086	1.5085	1.5085	1.5085	1.5085	1.5085
30	1.5144	1.5132	1.5128	1.5126	1.5123	1.5121	1.5121	1.5121	1.5120	1.5120	1.5120	1.5120	1.5120
40	1.5177	1.5165	1.5161	1.5159	1.5156	1.5154	1.5154	1.5154	1.5153	1.5153	1.5153	1.5153	1.5153
50	1.5210	1.5198	1.5194	1.5192	1.5189	1.5187	1.5187	1.5187	1.5186	1.5186	1.5186	1.5186	1.5186
60	1.5243	1.5231	1.5227	1.5225	1.5222	1.5220	1.5220	1.5220	1.5219	1.5219	1.5219	1.5219	1.5219
70	1.5277	1.5265	1.5261	1.5259	1.5256	1.5254	1.5254	1.5254	1.5253	1.5253	1.5253	1.5253	1.5253
80	1.5310	1.5298	1.5294	1.5292	1.5289	1.5287	1.5287	1.5287	1.5286	1.5286	1.5286	1.5286	1.5286

TABLE IV
ENTHALPY $H - H_0/RT_0$ FOR ARGON

$T, ^\circ\text{C.}$	0	25	50	75	100	125	150	174	200	300	400	500	600
P atm.													
0	0.0040	0.2328	0.4616	0.6904	0.9192	1.1480	1.3768	1.5965	1.8344	2.7496	3.6649	4.5801	5.4953
1	0.0000	0.2294	0.4587	0.6878	0.9170	1.1461	1.3751	1.5950	1.8331	2.7489	3.6646	4.5800	5.4950
2	-0.0041	0.2260	0.4557	0.6853	0.9147	1.1441	1.3734	1.5935	1.8318	2.7482	3.6644	4.5800	5.4948
5	-0.0160	0.2157	0.4469	0.6776	0.9081	1.1383	1.3684	1.5892	1.8279	2.7461	3.6636	4.5799	5.4940
10	-0.0298	0.1968	0.4303	0.6649	0.8970	1.1288	1.3600	1.5819	1.8215	2.7427	3.6624	4.5798	5.4928
20	-0.0598	0.1623	0.4032	0.6383	0.8770	1.1095	1.3423	1.5637	1.8088	2.7369	3.6599	4.5795	5.4897
30	-0.1136	0.1322	0.3750	0.6151	0.8526	1.0922	1.3252	1.5537	1.7993	2.7324	3.6574	4.5792	5.4877
40	-0.1517	0.0996	0.3470	0.5908	0.8324	1.0729	1.3114	1.5400	1.7844	2.7282	3.6549	4.5790	5.4851
50	-0.1893	0.0675	0.3194	0.5669	0.8117	1.0549	1.2968	1.5266	1.7725	2.7173	3.6525	4.5787	5.4826
60	-0.2262	0.0360	0.2923	0.5433	0.7912	1.0373	1.2805	1.5134	1.7610	2.7116	3.6500	4.5784	5.4801
70	-0.2626	0.0048	0.2655	0.5201	0.7711	1.0200	1.2656	1.5006	1.7498	2.7061	3.6475	4.5781	5.4775
80	-0.2985	-0.0259	0.2393	0.4974	0.7515	1.0029	1.2509	1.4881	1.7388	2.7010	3.6450	4.5778	5.4750

TABLE V
ENTROPY $S - S_0/R$ FOR ARGON

P atm.	$T, ^\circ\text{C.}$	0	25	50	75	100	125	150	174	200	300	400	500	600
0	∞	∞	∞	∞	∞	∞	∞	∞	∞	∞	∞	∞	∞	∞
1	0	-0.000	0.220	0.421	0.608	0.781	0.945	1.096	1.234	1.376	1.855	2.257	2.603	2.908
2	1	-0.686	-0.475	-0.274	-0.086	0.087	0.250	0.402	0.540	0.682	1.161	1.563	1.910	2.215
5	2	-1.622	-1.399	-1.197	-1.008	-0.834	-0.670	-0.518	-0.379	-0.237	-0.243	-0.046	0.993	1.298
10	5	-2.330	-2.104	-1.900	-1.710	-1.535	-1.370	-1.217	-1.077	-0.935	-0.453	-0.049	0.298	0.604
20	10	-3.052	-2.822	-2.613	-2.420	-2.243	-2.076	-1.921	-1.780	-1.637	-1.151	-0.746	-0.397	-0.091
30	20	-3.637	-3.407	-3.197	-2.999	-2.813	-2.644	-2.487	-2.343	-2.201	-1.696	-1.285	-0.906	-0.499
40	30	-4.064	-3.834	-3.624	-3.426	-3.240	-3.071	-2.914	-2.770	-2.627	-2.106	-1.690	-1.296	-0.885
50	40	-4.456	-4.226	-4.016	-3.818	-3.632	-3.463	-3.306	-3.162	-3.019	-2.488	-2.067	-1.677	-1.262
60	50	-4.810	-4.580	-4.370	-4.172	-3.986	-3.817	-3.659	-3.515	-3.372	-2.841	-2.414	-2.020	-1.603
70	60	-5.149	-4.919	-4.709	-4.511	-4.325	-4.156	-3.998	-3.854	-3.711	-3.180	-2.753	-2.357	-1.940
80	80	-5.461	-5.231	-5.021	-4.823	-4.637	-4.468	-4.310	-4.166	-4.023	-3.492	-3.065	-2.669	-2.252

TABLE VI
JOULE-THOMSON COEFFICIENT μ/μ_0 FOR ARGON

P atm.	$T, ^\circ\text{C.}$	0	25	50	75	100	125	150	174	200	300	400	500	600
0	∞	∞	∞	∞	∞	∞	∞	∞	∞	∞	∞	∞	∞	∞
1	0	1.000	0.854	0.737	0.642	0.559	0.484	0.422	0.368	0.326	0.178	0.062	-0.0070	-0.063
2	1	0.995	0.852	0.733	0.637	0.557	0.482	0.420	0.365	0.324	0.177	0.062	-0.0070	-0.063
5	2	0.991	0.849	0.731	0.635	0.557	0.482	0.420	0.365	0.324	0.176	0.062	-0.0070	-0.063
10	5	0.989	0.847	0.729	0.633	0.555	0.480	0.418	0.363	0.322	0.174	0.062	-0.0070	-0.063
20	10	0.989	0.847	0.729	0.633	0.555	0.480	0.418	0.363	0.322	0.174	0.062	-0.0070	-0.063
30	20	0.987	0.845	0.727	0.631	0.553	0.478	0.416	0.361	0.320	0.172	0.061	-0.0069	-0.063
40	30	0.984	0.842	0.724	0.628	0.550	0.475	0.413	0.358	0.317	0.169	0.061	-0.0069	-0.063
50	40	0.984	0.842	0.724	0.628	0.550	0.475	0.413	0.358	0.317	0.169	0.061	-0.0069	-0.063
60	50	0.982	0.840	0.722	0.626	0.548	0.473	0.411	0.356	0.315	0.167	0.061	-0.0069	-0.063
70	60	0.979	0.837	0.719	0.623	0.545	0.470	0.408	0.353	0.312	0.165	0.061	-0.0069	-0.063
80	80	0.977	0.835	0.717	0.621	0.543	0.468	0.406	0.351	0.310	0.163	0.061	-0.0069	-0.063

The properties are listed in dimensionless quantities. The conversion factors for various units are given in Table VII. In Fig. 1 the calculated values of μ are compared with the experimental values of Roebuck and Osterberg (3). The agreement is within the experimental error.

TABLE VII
CONVERSION FACTORS

To convert tabulated values of	to	with dimensions	multiply by
$\frac{PV}{P_0 V_0}$	PV	amagat atm. liter atm. mole ⁻¹ liter atm. gm. ⁻¹ cu. ft. atm. (lb. mole) ⁻¹ cu. ft. atm. lb. ⁻¹ cu. ft. p.s.i.a. (lb. mole) ⁻¹ cu. ft. p.s.i.a. lb. ⁻¹	1.00000 22.3919 0.560582 358.698 8.98002 5271.42 131.970
$\frac{C_p, C_v, S-S_0}{R \quad R \quad R}$	$C_p, C_v, S-S_0$	cal. mole ⁻¹ °C. ⁻¹ cal. gm. ⁻¹ °C. ⁻¹ j. mole ⁻¹ °C. ⁻¹ j. gm. ⁻¹ °C. ⁻¹ B.t.u. (lb. mole) ⁻¹ °F. ⁻¹ B.t.u. lb. ⁻¹ °F. ⁻¹	1.98719 0.0497494 83.1438 0.208151 1.98588 0.0497166
$\frac{H-H_0}{RT_0}$	$H-H_0$	cal. mole ⁻¹ cal. gm. ⁻¹ j. mole ⁻¹ j. gm. ⁻¹ B.t.u. (lb. mole) ⁻¹ B.t.u. lb. ⁻¹	542.821 13.5896 2271.17 56.8589 976.437 24.4451
$\frac{\mu}{\mu_0}$	μ	°C. atm. ⁻¹ °C. p.s.i.a. ⁻¹ °F. atm. ⁻¹ °F. p.s.i.a. ⁻¹	0.438 0.0298 0.788 0.0536

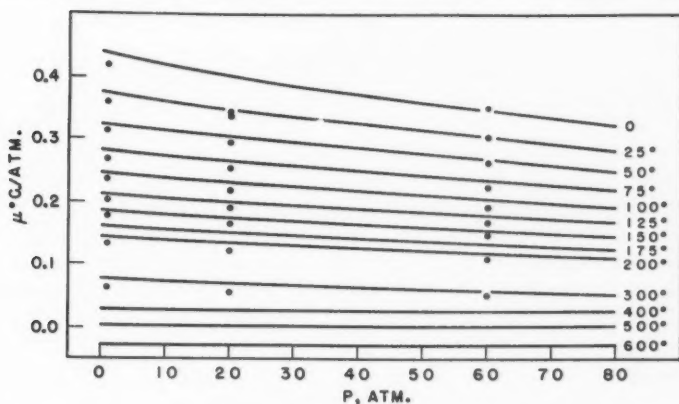


FIG. 1. Joule-Thomson coefficient vs. pressure at various temperatures. The curves are calculated and the points are the experimental values of Roebuck and Osterberg (3).

ACCURACY OF THE TABLES

It is not easy to state precisely the accuracy of the various quantities listed in the tables since apart from that of PV it depends on the accuracy with which the temperature derivatives of the virial coefficients can be estimated. The PV values are accurate to a few parts in 10^5 . The pressure variations of the other properties are probably accurate to a few per cent.

ACKNOWLEDGMENT

We wish to thank Mr. W. A. Evans for assistance in the computation of these tables.

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A HIGH-TEMPERATURE X-RAY GONIOMETER¹

BY R. G. BUTTERS² AND J. GORDON PARR³

ABSTRACT

A unit is described which, used in conjunction with a Philips high-angle X-ray diffraction goniometer, allows spectrometer plots to be made up to a temperature of 1100°C.

INTRODUCTION

The crystal structures of phases existing at elevated temperatures may only be properly determined by high-temperature X-ray analysis. While high-temperature diffraction cameras have been in use for many years, high-temperature attachments for Geiger-counter spectrometers are comparatively rare. The great advantage of the spectrometer over the camera is that continuous scans may be made at temperature during the course of a phase transformation, and relative quantities of phases present may be assessed directly from the spectrometer plot.

The unit to be described was made to fit a Philips high-angle X-ray diffraction goniometer, in which the specimen holder lies in a horizontal plane. Several attempts were made over the past few years to construct a suitable high-temperature unit to surround the specimen plate, but many difficulties were encountered, chief among which was the attainment of uniform specimen temperature. The apparatus finally designed has proved satisfactory in making structure analyses at temperatures up to 1100°C. It should be mentioned that the unit was made of materials that were readily available, and many dimensions are fairly flexible.

The features that are considered of greatest importance are:

1. The window arrangement to permit free entry and exit of the X-ray beam;
2. The nature of the furnace winding;
3. The aligning adjustment;
4. The general gas-tightness of the unit.

CONSTRUCTION

The high-temperature attachment is composed of three major parts, which are shown in Fig. 2.

A. A water-cooled specimen-holder which contains the gas-inlet, thermocouple leads, and a means of holding the specimen plate.

B. A water-cooled outer container, with gas-outlet, furnace terminal, and window.

C. A platinum-wound furnace, which fits semipermanently inside the outer container.

¹Manuscript received October 25, 1954.

Contribution from the Department of Mining and Metallurgy, The University of British Columbia, Vancouver, British Columbia.

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The general construction of the unit is shown in Fig. 1. The specimen-holder is fabricated from brass plate and rod with silver-soldered joints. A water-cooling coil is soft-soldered to this component (see Fig. 3). The specimen plate is of fused quartz approximately $\frac{5}{8}$ in. \times $\frac{1}{8}$ in. \times 3 in., attached to the semicircular end of the axial mounting shaft by a small screw. The powder specimen is placed on a thin molybdenum sheet which lies on the quartz plate. In order that the specimen-holder may be accurately indexed to the goniometer

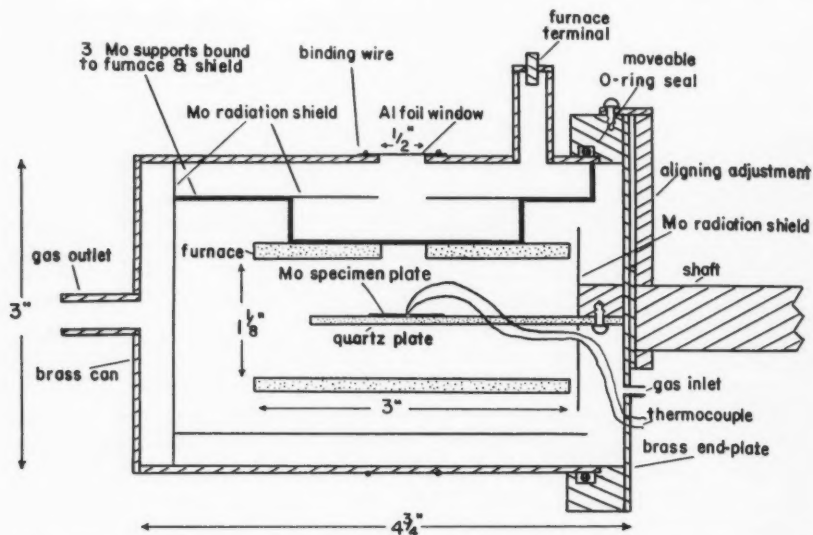


FIG. 1. High-temperature attachment. Approximately to scale. (Furnace winding and water-cooling are not shown.)

an adjustable clamp is mounted over the main mounting shaft and lies between the goniometer and the specimen holder (see Figs. 1 and 2). The clamp has a $\frac{1}{8}$ in. slot which engages with the indexing pin of the goniometer. Once the specimen-holder has been adjusted so that the plane of the quartz plate lies in the 0° plane of the goniometer, the indexing clamp is locked to the specimen holder by a small screw. Gas entry is made through the specimen-holder by a small brass tube; and thermocouple wires are led in via a small rubber bung. The thermocouple itself is made of 0.006 in. diameter Pt - Pt 10% Rh wire, and the junction rests on the molybdenum plate. Alternatively, each component of the couple may be spot-welded to either side of the molybdenum plate. A molybdenum radiation shield is fastened to the inside of the specimen-holder (see Figs. 1 and 3).

The outer can is made of 3 in. brass tubing and $\frac{1}{8}$ in. plate, and makes a vacuum-tight joint with the specimen-holder by means of a $\frac{1}{8}$ in. O-ring. The closed end of the can is provided with a gas-outlet. Near the open end of the can, a connection for one of the power leads is made through a rubber bung

held in a small piece of tubing (see Fig. 1). The second power lead is grounded to the can. The window is a semicircumferential $\frac{1}{2}$ in. slot cut out of the can and covered with a layer of 0.001 in. aluminum foil (Foil Wrap) sealed on by cellulose tape and secured by two binding wires pulling into circumferential grooves. Under vacuum the window sucks in about $\frac{1}{8}$ in., but breaks very rarely (see Fig. 4). The can is cooled by a few turns of flattened $\frac{3}{16}$ in. copper tubing soft-soldered in position.

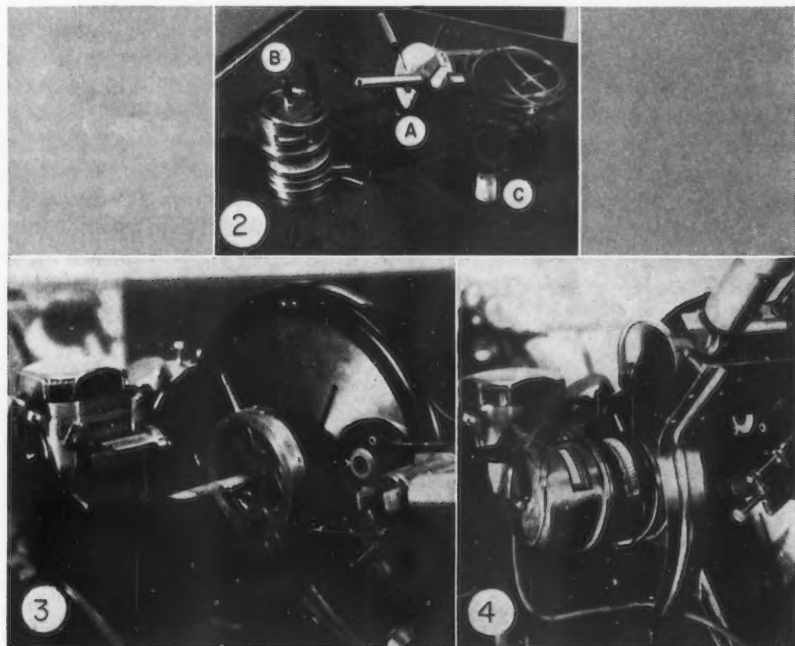


FIG. 2. Principal components of high-temperature attachment. FIG. 3. Specimen-holder in position in goniometer head. FIG. 4. High-temperature attachment in position.

The furnace is made from a piece of vitreous silica tubing, 3 in. long, $1\frac{1}{8}$ in. I.D., with a semicircumferential slot to coincide radially with the window in the outer can. Each end of the tube has 36 radial slots cut in it to hold the platinum winding which runs longitudinally inside the tube. The method of making the winding may be seen in Fig. 2.

The furnace tube is supported coaxially in the can by three stout molybdenum wires, which also support a $2\frac{3}{8}$ in. diameter cylindrical molybdenum radiation shield. The supporting wires are attached at the end furthest from the specimen-holder to a radiation shield that fits snugly in the outer can; the other ends of the wires are sprung against the outer can (see Fig. 1). The furnace, radiation shield, and window are, of course, placed so that their circumferential slots are aligned in the plane of rotation of the goniometer.

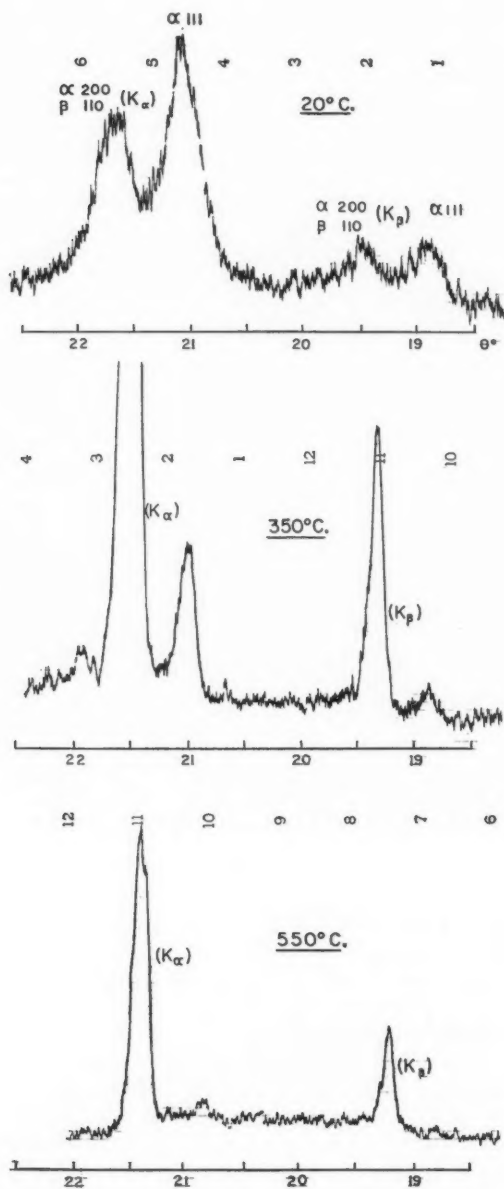


FIG. 5. Diffraction plots on Cu-Zn alloy. (Cu K radiation.)

OPERATION

The powder specimen is placed on the molybdenum specimen plate, and the thermocouple junction is adjusted so that it dips into the powder. The can, with furnace and radiation shield, is pushed into place. Cooling water is circulated, and the unit is evacuated. Helium, purified by passage through activated silica gel at liquid air temperature, is passed through the can for two or three hours before the furnace is switched on.

The temperature is controlled manually by a variable transformer. No difficulty is experienced in keeping the temperature constant to within $2^{\circ}\text{C}.$, so long as the cooling-water supply is steady. Some difficulty has been encountered in keeping highly reactive metals, like titanium, free from oxidation at high temperatures, but in such cases the specimen is mixed with zirconium powder which acts as a getter. It is believed that the source of oxygen is the vitreous silica furnace-tube, which, apparently, cannot be adequately degassed.

Fig. 5 shows plots made of a 45% zinc - 55% copper alloy at three temperatures. Copper K radiation was used. At $20^{\circ}\text{C}.$ the powder showed broad lines (due to internal strain produced by filing) of 111α (face-centered cubic) and the coincident 200α and 110β (body-centered cubic). Both $K\alpha$ and $K\beta$ peaks were produced as the radiation was not filtered. At $350^{\circ}\text{C}.$ the 111α peak is considerably reduced in intensity; and at $550^{\circ}\text{C}.$ it has almost disappeared. The phase diagram for copper-zinc implies that the $(\alpha+\beta) - \beta$ boundary should be crossed about $500^{\circ}\text{C}.$

The plots also show that the diffraction lines are intense, despite absorption by the foil window and possibly by the platinum furnace wires. Further, the shift in angle as the cell parameter changes with temperature is clearly indicated.

ACKNOWLEDGMENT

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THE PENETRATION OF CONCRETE BY RADIONUCLIDES IN SOLUTION¹

By L. H. GEVANTMAN AND D. SAM

ABSTRACT

The penetration of concrete by the Cs^{134} , Nb^{95} , Ru^{106} , Ce^{144} , and I^{131} in solution is measured. Diffusion constants for Cs^{134} and I^{131} are calculated. The values obtained are compared with those measured by Spinks in neat cement and mortar.

INTRODUCTION

The widespread use of radioactive elements in research and industry creates an increased frequency in the contamination of surfaces which fix undesirable quantities of radioactivity. Another serious contamination problem arises when high concentrations of products from the detonation of nuclear weapons contact and adhere to the outer surfaces of structures.

Concrete is representative of a group of materials which not only absorb large quantities of radionuclides but also resist their removal by common cleaning methods. When it is realized that concrete comprises a large percentage of the exposed surfaces in structures it becomes clear that more information about the contamination process is needed. A limited contribution to this problem is made by the present study of the depths to which specific radionuclides in solution penetrate concrete.

Diffusion of radionuclides in solids has been measured in lead by Groh and Hevesy (2) and in gels by Salvinien, Marignan, and Cordier (3). Spinks, Baldwin, and Thorvaldson (4) measured the penetration of neat cement and mortar by solutions of Na_2SO_4 , CaSO_4 , and NaI which contained Na^{22} , Ca^{45} , I^{131} , and S^{35} to trace the ions of interest. On the basis of these measurements they calculated a diffusion constant for each ion to describe its observed rate of penetration.

This study has extended the work of Spinks *et al.* to concrete. In contrast to their direct measurement of concentration gradients by surface β -counting at constant geometry this method measured the variation in concentration with depth by observing the decrease in γ activity associated with the removal of a thin section of solid. The activity contained in each section was used to obtain concentration gradients from which the diffusion constants for two radioelements were calculated.

EXPERIMENTAL DETAILS

On a dry basis each pound of concrete consisted of $\frac{1}{8}$ lb. Portland cement, $\frac{1}{8}$ lb. sand, and $\frac{1}{2}$ lb. aggregate. Approximately $\frac{1}{10}$ lb. of water was added to slurry the above mixture and additional water was used to cure the solid

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after it was formed. The concrete was cast in blocks $1\frac{1}{4}$ in. by $1\frac{1}{4}$ in. by 1 in. which were coated with paraffin wax on all but the evenly ground top surface and were immersed for a definite time in approximately 0.5 liters of solution containing the elements of interest. Following exposure, the sample was removed, dried, counted, and then sectioned. Sectioning was accomplished by grinding the sample surface on the face of a rotating abrasive cutting wheel. This operation was performed in a dry box to reduce the activity inhalation hazard. The sample was counted through a 720 mgm. per sq. cm. aluminum absorber on a gas-flow proportional counter. An activity measurement was made after each section and an activity gradient obtained. The sample was measured in a fixed position during the entire series of activity readings to avoid geometry corrections and employment of γ -counting avoided the necessity of correcting for absorption.

The solution to Fick's law of diffusion (1) for the case of a semi-infinite solid is used to compute the diffusion constant. Thus,

$$[1] \quad 1 - \frac{C}{C_0} = \frac{2}{\sqrt{\pi}} \int_0^\beta e^{-y^2} dy.$$

In applying equation 1 to this problem, it is assumed that the activity measured at each layer is proportional to the concentration C at depth x (cm.) and time t (sec.). Also it is assumed that for all values of t at $x = 0$, $C_0 = \sum C$. In equation 1, β is defined by:

$$[2] \quad \beta = \frac{x}{2\sqrt{Dt}}$$

where D is the diffusion constant. From tables (5) may be read values of β vs. the integral

$$\frac{2}{\sqrt{\pi}} \int_0^\beta e^{-y^2} dy.$$

From equation 2,

$$[3] \quad D = \frac{1}{4t} \left(\frac{x}{\beta} \right)^2.$$

RESULTS

The results shown in Table I represent a typical set of experimental data and calculations. A plot of the data showing β vs. x is given in Fig. 1. The diffusion constants which are calculated for Cs and I at two concentrations are given in Table II. A comparison is made in Table III between these values and those measured by Spinks for cement and mortar.

General agreement exists between the results observed for cement and mortar and those for concrete. The diffusion constant measured in concrete varies with time of immersion but tends to approach a limiting value as the time is increased. The constant for the anion is larger than that of the cation by a factor of approximately 10^2 .

TABLE I
TYPICAL EXPERIMENTAL DATA AND CALCULATIONS FOR 0.01 *N* KI CONTAINING I^{131} ^a

Experimental data		Calculations		
x (cm.)	Activity, c.p.m. $\times 10^{-3}$	x (cm.)	C/C_0	β
0	25.3	0	1	0
0.031	24.41	0.1	0.85	0.1338
0.193	16.35	0.2	0.68	0.2917
0.557	7.48	0.3	0.505	0.4714
0.766	5.28	0.4	0.415	0.5764
1.017	2.58	0.5	0.338	0.6775
		0.6	0.27	0.7800

^aSolution was made 0.01 *N* in HSO_3^- ion to hold the iodine as I^- .

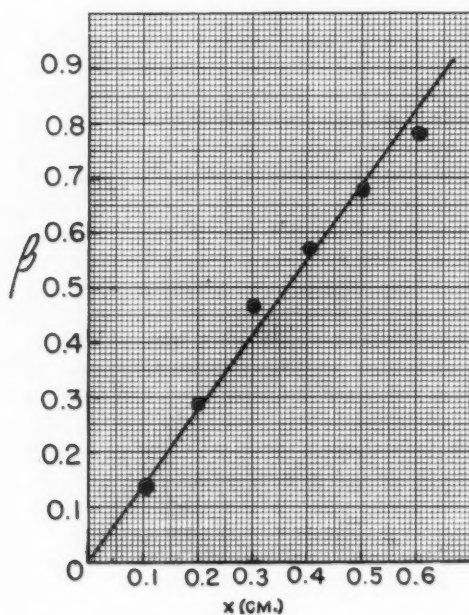


FIG. 1. β vs. x from data for I^{131} in solution 0.01 *N* KI and 0.01 *N* $NaHSO_3$.

TABLE II
DIFFUSION CONSTANTS FOR Cs^{134} AND I^{131} AS A FUNCTION OF CONCENTRATION AND TIME

Time (days)	$C_I = 8 \times 10^{-13} M$	$C_I = 0.01 M$	$C_{Cs} = 3 \times 10^{-9} M$	$C_{Cs} = 0.01 M$
	Diffusion constants D (sq. cm./sec. $\times 10^{-10}$)			
1	7930	15,100	28	213
3	1690	5050	19.7	53.9
5	1580	3030	24.5	20.8

TABLE III

COMPARISON OF DIFFUSION CONSTANTS FOR CONCRETE, NEAT CEMENT, AND MORTAR

Radionuclide	Solution concentration	Neat cement ^a	Mortar ^a (1:2)	Concrete ^b
		Diffusion constants (sq. cm./sec. $\times 10^{-10}$)		
I ¹³¹	8×10^{-13} M KI	—	—	1580
I ¹³¹	0.01 M KI	—	—	3020
I ¹³¹	0.15 M NaI	—	130	—
Cs ¹³⁴	3×10^{-9} M CsCl	—	—	24.5
Cs ¹³⁴	0.01 M CsCl	—	—	20.8
Na ²²	0.15 M Na ₂ SO ₄	—	3.0	—
Ca ⁴⁵	Saturated CaSO ₄	0.1	2.8	—

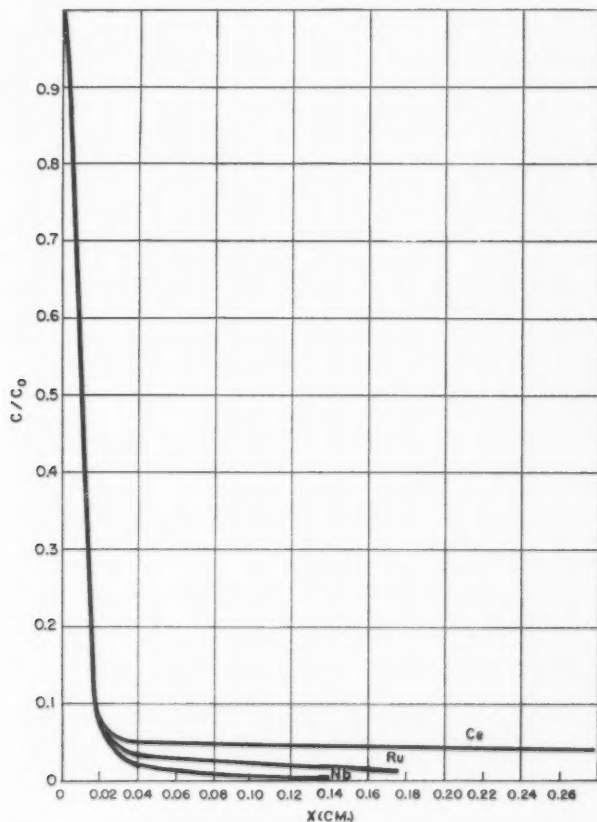
^aValues obtained from Spinks (3).^bConstants for concrete measured at five days and constants for cement and mortar measured at four weeks.

FIG. 2. Activity distribution of Ru¹⁰⁶, Nb⁹⁵, and Ce¹⁴⁴ in concrete after five days. Initial concentration: Nb⁹⁵— 9.6×10^{-9} M; Ru¹⁰⁶— 4.9×10^{-8} M; Ce¹⁴⁴— 2.8×10^{-8} M.

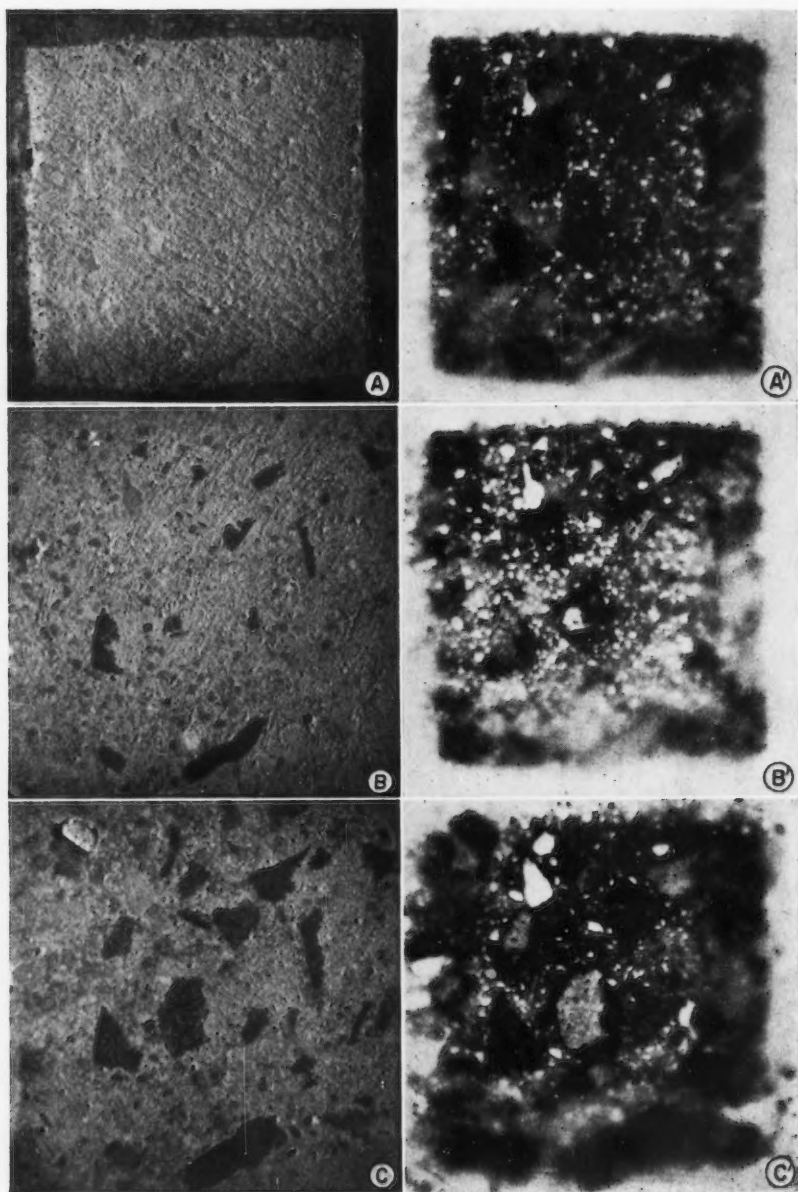


FIG. 3. Typical photomicrographs and matching autoradiographs ($4\times$ magnification) of concrete block surfaces before and after sectioning. A,A': top surface; B,B': first section; C,C': second section.

Concentration of the solution does not alter the value of the constant but does enable the limiting value to be approached more rapidly. The increase in the diffusion constant of I by a factor of 10 in going from mortar to concrete may simply be a measure of the increase in the porosity of the solid. Similarly the increase in constant for Ca in cement and mortar may be explained on this basis. An alternative explanation for the increase in value observed between Cs and Na may be found in the lower degree of hydration of the cesium ion despite an increase in its ionic radius.

In Fig. 2 a group of curves showing the activity distribution of Ru^{106} , Nb^{95} , and Ce^{144} in concrete are given. They all show a small depth of penetration; for this reason their diffusion constants were not calculated. Fig. 3 gives a matching set of photographs and autoradiographs of the concrete surface before and after sectioning. It is possible in this way to follow the path of activity through the solid.

DISCUSSION

This experiment permits the measurement of a rate of penetration into concrete by the radionuclides examined. This measurement in turn leads to a qualitative estimate of the extent of contamination of concrete. Because of the composition of the solutions employed, the results are more easily applied to the industrial research situation where proportionately large concrete areas are involved.

The results extend the work by Spinks and show agreement with it. Generally it may be concluded that anions penetrate concrete at a greater rate and to greater depth than do cations. There appears to be very little difference in penetration rates between soluble cations. It also seems that the trend of porosity in these solids is neat cement < mortar < concrete.

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